



INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : C07D 403/10, C08K 5/3475	A1	(11) International Publication Number: WO 99/23093 (43) International Publication Date: 14 May 1999 (14.05.99)
<p>(21) International Application Number: PCT/EP98/06654</p> <p>(22) International Filing Date: 22 October 1998 (22.10.98)</p> <p>(30) Priority Data: MI97A002436 30 October 1997 (30.10.97) IT</p> <p>(71) Applicant (for all designated States except US): GREAT LAKES CHEMICAL ITALIA S.R.L. [IT/IT]; Via Cornaggia, 10, I-20123 Milano (IT).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): RIVA, Rosa, Maria [IT/IT]; Via S. Protasio, 11, I-Merate (IT). NERI, Carlo [IT/IT]; Via Europa, 32, I-20097 S. Donato M.se (IT). COLOMBO, Rosalba [IT/IT]; Via Manzoni, 12, I-Vimercate (IT).</p> <p>(74) Agents: FRANCK, Peter et al.; Uexküll & Stolberg, Beselerstrasse 4, D-22607 Hamburg (DE).</p>		<p>(81) Designated States: CA, JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).</p> <p>Published <i>With international search report.</i></p>
<p>(54) Title: 2-(2'-HYDROXYPHENYL)BENZOTRIAZOLES USED AS U.V. STABILISERS</p> <div data-bbox="500 1144 1112 1407" data-label="Chemical-Block"> <p style="text-align: right;">(I)</p> </div> <p>(57) Abstract</p> <p>2-(2'-hydroxyphenyl)benzotriazoles having general formula (I). The above 2-(2'-hydroxyphenyl)benzotriazoles are useful as light stabilizers for organic polymers.</p>		

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2-(2'-HYDROXYYPHENYL)BENZOTRIAZOLES USED AS U.V. STABILISERS

The present invention relates to 2-(2'-hydroxyphenyl)benzotriazoles.

5 More specifically, the present invention relates to 2-(2'-hydroxyphenyl)benzotriazoles containing a 2,4-imidazolidinedione group or a 2,4-imidazolidine-dione-5,5-disubstituted group in the molecule, a process for their preparation and their use as light
10 stabilizers for organic polymers.

The present invention also relates to the polymeric compositions stabilized with the above benzotriazoles and to the end-products obtained from these compositions.

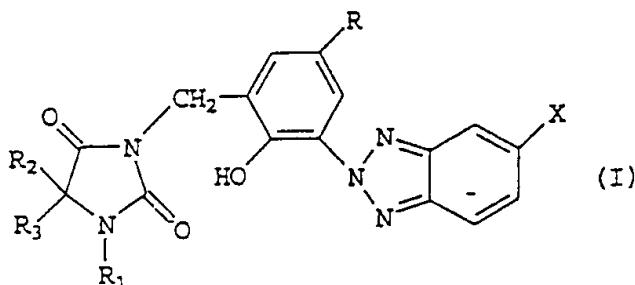
15 2-(2'-hydroxyphenyl)benzotriazoles which can be used as light stabilizers are known in the art. These benzotriazoles however have various disadvantages. In fact, they are often rather volatile, they have a low thermal stability and, as they sometimes have a signi-

ficant absorption at 400 nm, they give the polymers, into which they are incorporated, a yellow colouring.

The Applicant has now surprisingly found that 2-(2'-hydroxyphenyl)benzotriazoles containing a 2,4-
 5 imidazolidinedione group or a 2,4-imidazolidinedione-5,5-disubstituted group in the molecule, are capable of overcoming the drawbacks of the known art. In fact, the above benzotriazoles have a low volatility (they are therefore able to remain inside the stabilized organic
 10 polymer for a longer period) and also a high thermal stability. In addition, they have a low absorption at $\lambda = 400$ nm and maintain the absorption at the two λ typical of benzotriazoles, at about 300 nm and 340 nm and consequently they do not give a yellow colouring to
 15 the polymers into which they are incorporated.

The present invention therefore relates to 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I):

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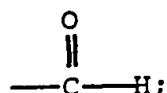


wherein:

25 - X represents a hydrogen atom; a halogen atom

selected from chlorine and bromine; a linear or branched C_1-C_{18} alkyl group; a linear or branched C_1-C_{18} alkoxy group; a cyano group;

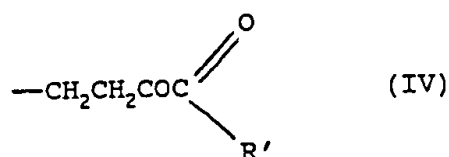
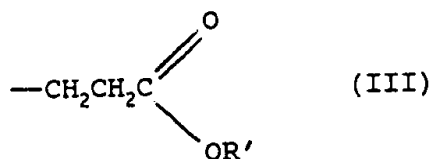
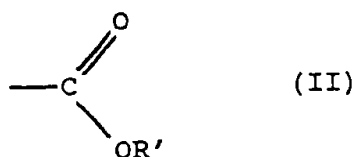
- R represents a halogen atom selected from chlorine and bromine; a linear or branched C_1-C_{18} alkyl group; a linear or branched C_2-C_{18} alkenyl group; a linear or branched C_2-C_{18} alkynyl group; a C_5-C_{18} cycloalkyl group, said cycloalkyl group optionally substituted; a C_7-C_{15} arylalkyl or alkylaryl group; a C_6-C_{14} aryl group, said aryl group optionally substituted; a linear or branched C_1-C_{18} alkoxy group; a heterocyclic group with 5 or 6 atoms containing at least one heteroatom selected from oxygen, nitrogen and sulfur, said heterocyclic group optionally substituted; a group having the formula:



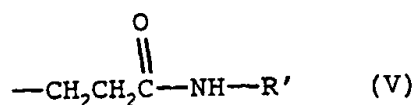
- a $-\text{COR}_4$ group or a $-\text{NR}_5\text{R}_6$ group wherein R_4 , R_5 and R_6 , the same or different, represent a linear or branched C_1-C_{18} alkyl group; a linear or branched C_2-C_{18} alkenyl group; a linear or branched C_2-C_{18} alkynyl group; a C_5-C_{18} cycloalkyl group, said cycloalkyl group optionally substituted; a C_7-C_{15} arylalkyl or alkylaryl group; a C_6-C_{14} aryl group,

said aryl group optionally substituted; a hetero-
cyclic group with 5 or 6 atoms containing at least
one heteroatom selected from oxygen, nitrogen and
sulfur, said heterocyclic group optionally substi-
tuted;

or R represents an ester group having general
formula (II), (III) or (IV):

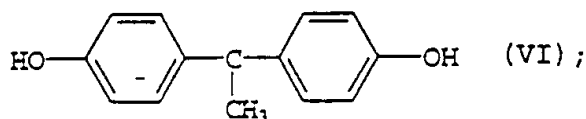


or an amide group having general formula (V):



wherein R' represents a linear or branched C₁-C₁₈
alkyl group; a linear or branched C₂-C₁₈ alkenyl
group; a linear or branched C₂-C₁₈ alkynyl group;
a C₅-C₁₈ cycloalkyl group, said cycloalkyl group
optionally substituted; a C₇-C₁₅ arylalkyl or
alkylaryl group; a C₆-C₁₄ aryl group, said aryl

group optionally substituted; a linear or branched
 C_1-C_{18} alkoxyl group; a heterocyclic group with 5
 or 6 atoms containing at least one heteroatom
 selected from oxygen, nitrogen and sulfur, said
 5 heterocyclic group optionally substituted;
 or R represents a 4,4'-ethylidenebisphenol group
 having formula (VI):



10

- R_1 represents a hydrogen atom; a linear or
 branched C_1-C_{18} alkyl group; a linear or branched
 C_2-C_{18} alkenyl group; a linear or branched C_2-C_{18}
 alkynyl group; a C_5-C_{18} cycloalkyl group, said
 15 cycloalkyl group optionally substituted; a C_7-C_{15}
 arylalkyl or alkylaryl group; a C_6-C_{14} aryl group,
 said aryl group optionally substituted; a hetero-
 cyclic group with 5 or 6 atoms containing at least
 one heteroatom selected from oxygen, nitrogen and
 20 sulfur, said heterocyclic group optionally substi-
 tuted; an acyl group having general formula (VII):



or an ester group having general formula (VIII):

25



wherein R' has the same meanings described above;

- R₂ and R₃, the same or different, represent a
 5 hydrogen atom; a linear or branched C₁-C₁₈ alkyl
 group; a phenyl group; a heterocyclic group with
 5 or 6 atoms containing at least one heteroatom
 selected from oxygen, nitrogen and sulfur, said
 heterocyclic group optionally substituted.

10 The compounds having general formula (I) can be
 used as light stabilizers for organic polymers.

When the C₃-C₁₈ cycloalkyl groups, the C₆-C₁₄ aryl
 groups and the heterocyclic groups with 5 or 6 atoms
 are defined as being optionally substituted, these
 15 groups are substituted with: halogen atoms selected
 from chlorine and bromine, linear or branched C₁-C₁₈
 alkyl groups, linear or branched C₂-C₁₈ alkenyl groups;
 linear or branched C₂-C₁₈ alkynyl groups, OH groups, NH
 groups, SH groups.

20 Examples of C₁-C₁₈ alkyl groups are: methyl, ethyl,
 propyl, isopropyl, n-butyl, sec-butyl, t-butyl, t-amyl,
 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-do-
 decyl, 1,1,7,7-tetramethyloctyl, n-octadecyl, etc.

Examples of C₂-C₁₈ alkenyl groups are: vinyl,
 25 propylene, butylene, pentylene, hexylene, etc.

Examples of C_2-C_{18} alkynyl groups are: acetylene, propyne, butyne, 2-butyne, etc.

Examples of C_5-C_{18} cycloalkyl groups, optionally substituted, are: cyclohexyl, cyclopentyl, methylcyclo-
5 hexyl, etc.

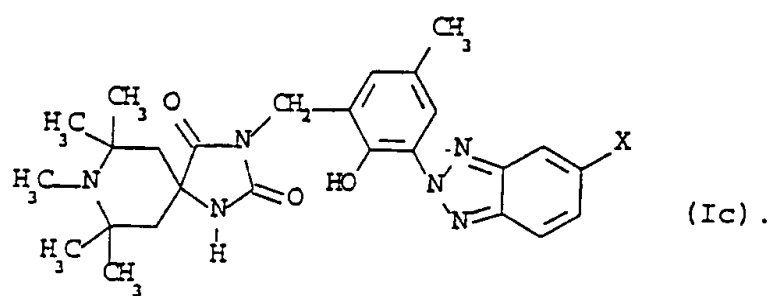
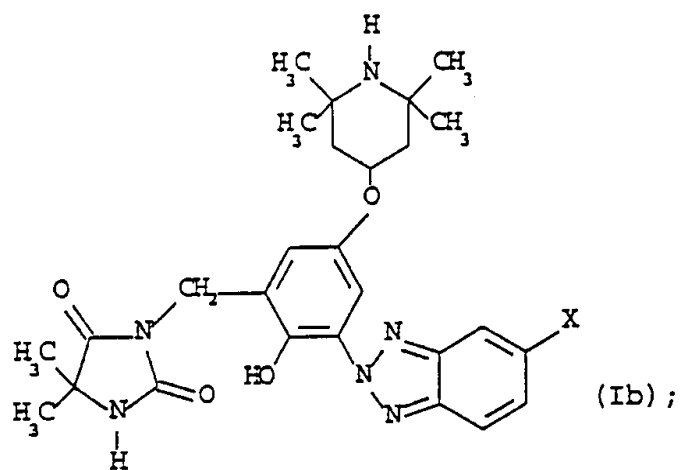
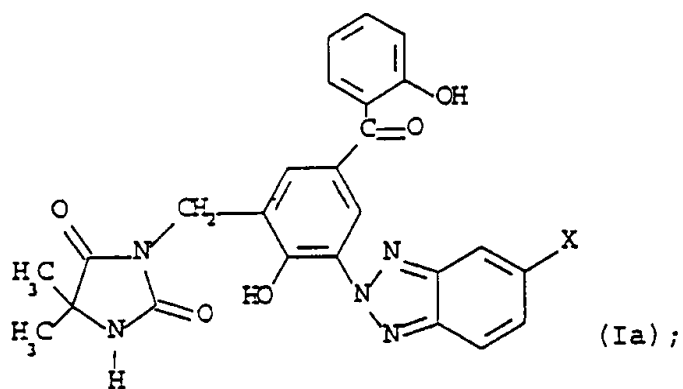
Examples of C_7-C_{15} arylalkyl or alkylaryl groups are: benzyl, 2-phenylethyl, 4-t-butylbenzyl, etc.

Examples of C_6-C_{14} aryl groups, optionally substituted, are: phenyl, naphthyl, anthracenyl, 2-hydroxy-
10 phenyl, etc.

Examples of C_1-C_{18} alkoxy groups are: methoxyl, ethoxyl, propoxyl, n-butoxyl, etc.

Examples of heterocyclic groups with 5 or 6 atoms, optionally substituted, are: piperidine, morpholine,
15 piperazine, triazole, tetramethylpiperidine, pentamethylpiperidine, tetramethylmorpholine, pentamethylmorpholine, 4-hydroxy-tetramethylpiperidine, etc.

Specific examples of compounds having general formula (I), which in no way limit the scope of the
20 present invention, are:

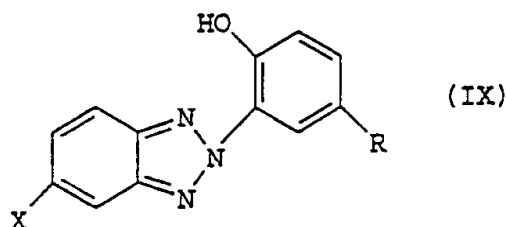


The compounds having general formula (I) of the present invention can be prepared with various processes.

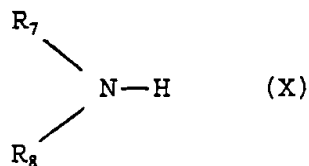
A further object of the present invention relates to a process for the preparation of 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I).

A process for the preparation of compounds having general formula (I) comprises:

- (a) reacting a 2-(2'-hydroxyphenyl)benzotriazole having general formula (IX):

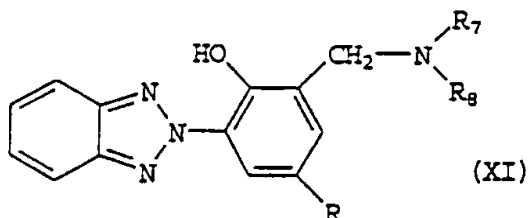


wherein X and R have the same meanings defined above, with a secondary amine having general formula (X):



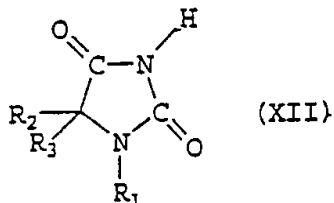
wherein R_7 and R_8 , the same or different, preferably the same, represent a linear alkyl group having from 3 or more carbon atoms, in the presence of formaldehyde or para-formaldehyde, preferably para-formaldehyde and an inert organic

solvent such as, for example, an alcohol, preferably n-butanol, at a temperature ranging from 95°C to 100°C, for a time ranging from 40 hours to 60 hours, obtaining the Mannich base having general formula (XI):



wherein R, R₇ and R₈ have the same meanings defined above;

(b) reacting the Mannich base having general formula (XI) obtained in step (a) with a 2,4-imidazolidinedione having general formula (XII):



wherein R₁, R₂ and R₃ have the same meanings described above, in the presence of an alkaline catalyst such as, for example, sodium methyrate, sodium hydroxide, potassium hydroxide, lithium hydroxide, etc., and an inert organic solvent having a boiling point at 160°C such as, for example, 1,2,4-trimethylbenzene, 4-isopropyl-

benzene, etc., at a temperature ranging from 20°C to 200°C, preferably between 40°C and 170°C, for a time ranging from 20 hours to 30 hours, obtaining a solution from which, after cooling to room temperature and eliminating the catalyst by filtration and the solvent by evaporation, the desired compound is isolated by crystallization in the presence of an inert organic solvent selected from alcohols (for example, methanol, etc.); monoalkylethers (for example, 2-methoxyethanol, etc.); linear or cyclic aliphatic hydrocarbons (for example, hexane, heptane, cyclohexane, methylcyclohexane, etc.); aromatic hydrocarbons (for example, toluene, etc.); chlorinated aromatic solvents (for example, chlorobenzene, etc.).

The 2-(2'-hydroxyphenyl)benzotriazoles having general formula (IX) can be prepared as described, for example, in German patent application DE 4.237.817.

The secondary amines having general formula (X) and 2,4-imidazolidinediones having general formula (XII), are commercially available compounds.

As already mentioned above, the compounds having general formula (I) of the present invention can be used as light stabilizers for a wide range of organic polymers.

Organic polymers capable of being stabilized with the compounds of the present invention are:

- (1) polymers of mono-olefins and diolefins such as, for example, polypropylene, polyisobutylene,
5 polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene; as well as polymers of cyclo-olefins such as, for example, cyclopentene or norbornene; polyethylene (which can be optionally cross-linked) such as, for example, high
10 density polyethylene (HDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), branched low density polyethylene (BLDPE).

Polyolefins such as, for example the monolefins
15 mentioned in the above paragraph, preferably polyethylene and polypropylene, can be prepared with various methods known in literature, preferably using the following methods:

- (a) radicalic polymerization (generally carried out at
20 a high pressure and high temperature);
- (b) catalytic polymerization using a catalyst which normally contains one or more metals of groups IVb, Vb, VIb or VIII of Periodic Table. These metals generally have one or more ligands such as,
25 for example, oxides, halides, alcoholates, ethers,

amines, alkyls, alkenyls and/or aryls which can be π - or σ -co-ordinated. These metal complexes can be in free form or supported in substrates such as, for example activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts can be soluble or insoluble in the reaction medium. The catalysts can be used alone or in the presence of other activators such as, for example, metal alkyls, metal hydrides, halides of metal alkyls, oxides of metal alkyls or metal alkyloxanes, these metals being elements belonging to groups Ia, IIa and/or IIIa of the Periodic Table. The activators can be conveniently modified with other ester, ether, amine or silyl-ether groups. These catalytic systems are usually called Phillips, Standard Oil Indiana, Ziegler(-Natta), TNZ (Du-Pont), metallocene or "single site catalyst" (SSC).

- (2) Mixtures of the polymers described under point (1) such as, for example, mixtures of polypropylene with polyisobutylene; mixtures of polypropylene with polyethylene (for example, PP/HDPE, PP/LDPE); mixtures of different types of polyethylene (for example, LDPE/HDPE).
- (3) Copolymers of mono-olefins and diolefins with each

other or with other vinyl monomers such as, for example, ethylene-propylene copolymers, linear low density polyethylene (LLDPE) and its mixtures with low density polyethylene (LDPE), propylene/but-1-ene copolymers, propylene/isobutylene copolymers, ethylene/but-1-ene copolymers, ethylene/hexene copolymers, ethylene/methylpentene copolymers, ethylene/heptene copolymers, ethylene/octene copolymers, propylene/butadiene copolymers, isobutylene/isoprene copolymers, ethylene/alkyl acrylate copolymers, ethylene/alkyl methacrylate copolymers, ethylene/vinyl acetate copolymers and their copolymers with carbon monoxide or ethylene/acrylic acid copolymers and their salts (ionomers) as well as terpolymers of ethylene with polypropylene and a diene such as, for example, hexadiene, dicyclopentadiene or ethylidene-norbornene; and mixtures of these copolymers with each other or with the polymers cited in paragraph (1) such as, for example, polypropylene/ethylene-propylene copolymers, LDPE/ethylene-vinylacetate (EVA) copolymers, LDPE/ethylene-acrylic acid (EAA) copolymers, LLDPE/EVA, LLDPE/EAA, and alternating or "random" polyalkylene/carbon monoxide copolymers and their mixtures with other polymers such

as, for example, polyamides.

- (4) Hydrocarbon resins (for example, C₅-C₉) comprising their hydrogenated modifications (for example, adhesive agents) and mixtures with polyalkylene and starch.
- (5) Polystyrene, poly(p-methylstyrene), poly(α -methylstyrene).
- (6) Copolymers of styrene or α -methylstyrene with dienes or acrylic derivatives such as, for example, styrene/butadiene, styrene/acrylonitrile, styrene/alkyl methacrylate, styrene/butadiene/alkyl acrylate, styrene/butadiene/alkyl methacrylate, styrene/maleic anhydride, styrene/acrylonitrile/methylacrylate; mixtures, having a high impact strength, between copolymers of styrene and another polymer such as, for example, a polyacrylate, a polymer of a diene or an ethylene/propylene/diene terpolymer, block polymers of styrene such as, for example, styrene/butadiene/styrene, styrene/isoprene/styrene, styrene/ethylene/butylene/styrene or styrene/ethylene/propylene/styrene.
- (7) Grafted copolymers of styrene or α -methylstyrene such as, for example, styrene in polybutadiene, styrene in polybutadiene or polybutadiene-acrylo-

- 5 nitrile copolymers; styrene and acrylonitrile (or methacrylonitrile) in polybutadiene; styrene, acrylonitrile and methylmethacrylate in polybutadiene; styrene and maleic anhydride in polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide in polybutadiene; styrene and maleimide in polybutadiene; styrene and alkylacrylates or methacrylates in polybutadiene; styrene and acrylonitrile in ethylene/propylene/diene terpoly-
- 10 mers, styrene and acrylonitrile in polyalkyl acrylates or polyalkyl methacrylates, styrene and acrylonitrile in acrylate/butadiene copolymers, as well as mixtures of the copolymers listed above with the copolymers cited under point (6) such as,
- 15 for example, mixtures of known copolymers such as ABS, MBS, ASA or AES;
- (8) Polymers containing halogens such as, for example, polychloroprene, chlorinated rubbers, chlorinated or chlorosulfonated polyethylene, ethylene and
- 20 chlorinated ethylene copolymers, homopolymers and copolymers of epichlorohydrin, in particular polymers of vinyl compounds containing halogens such as, for example, polyvinyl chloride, polyvinylidenechloride, polyvinyl fluoride or polyvinylidene fluoride; and also their copolymers such
- 25

as, for example, vinyl chloride/vinylidenechloride, vinyl chloride/vinyl acetate or vinylidenechloride/vinyl acetate.

- 5 (9) Polymers deriving from α,β -unsaturated acids and their derivatives such as, for example, polyacrylates and polymethacrylates, polymethyl methacrylates, polyacrylamides and polyacrylonitriles, modified with butyl acrylate.
- 10 (10) Copolymers of monomers according to point (9) with each other or with other unsaturated monomers such as, for example, acrylonitrile/butadiene copolymers, acrylonitrile/alkylacrylate copolymers, acrylonitrile/alkoxyalkyl acrylate copolymers or acrylonitrile/vinyl halide copolymers or acrylonitrile/alkyl methacrylate/butadiene terpolymers.
- 15 (11) Polymers deriving from unsaturated alcohols and amines, or their acyl or acetal derivatives such as, for example, polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyrral, polyallyl phthalate or polyallyl melamine; and also their copolymers with the olefins listed under point (1).
- 20 (12) Homopolymers and copolymers of open-chain ethers or cyclic ethers such as, for example, polyalkyle-

ne glycols, polyethyleneoxide, polypropylene oxide, or copolymers of the compounds described above with bis-glycidyl ethers.

- 5 (13) Polyacetals such as, for example, polyoxymethylene and polyoxymethylenes which contain ethylene oxide as comonomer; polyacetals modified with thermo-plastic polyurethanes, acrylates or MBS.
- 10 (14) Polyphenylene oxides and sulfides and mixtures of polyphenylene oxides with styrene or polyamide polymers.
- 15 (15) Polyurethanes deriving from hydroxyl-terminated polyethers, polyesters or polybutadienes on the one hand and aliphatic or aromatic polyisocyanates on the other, as well as the precursors of the above compounds.
- 20 (16) Polyamides and copolyamides deriving from diamines and dicarboxylic acids and/or aminocarboxylic acids or from the corresponding lactams such as, for example, polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12, 4/6, 12/12, polyamide 11, polyamide 12, aromatic polyamides obtained starting from m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic and/or terephthalic acid and with or
- 25 without an elastomer as modifier, for example,

poly-2,4,4-trimethylhexamethylene terephthalamide
or poly-m-phenylene isophthalamide; and also block
copolymers of the above polyamides with poly-
olefins, olefinic copolymers, ionomers or elasto-
5 mers chemically bound or grafted; or with poly-
ethers such as, for example, polyethylene glycol,
polypropylene glycol or polytetramethylene glycol;
as well as polyamides or copolyamides modified
with EPDM or ABS; and polyamides condensed during
10 processing ("RIM polyamide system").

(17) Polyureas, polyimides, polyamide-imides and
polybenzimidazoles.

(18) Polyesters deriving from dicarboxylic acids and
diols and/or from hydroxycarboxylic acids or from
15 the corresponding lactones such as, for example,
polyethylene terephthalate, polybutylene tereph-
thalate, poly-1,4-dimethylolcyclohexane terephtha-
late and polyhydroxybenzoates, as well as block
copolyether esters deriving from polyethers with
20 hydroxyl-terminated groups; and also polyesters
modified with polycarbonates or MBS.

(19) Polycarbonates and polyester carbonates.

(20) Polysulfones, polyethersulfones and polyetherketo-
nes.

25 (21) Cross-linked polymers deriving from aldehydes on

the one hand and from phenols, urea and melamines on the other, such as, for example, phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.

5 (22) Drying or non-drying alkyd resins.

(23) Resins based on unsaturated polyesters deriving from copolyesters of dicarboxyl acids saturated and unsaturated with polyhydric alcohols and vinyl compounds as cross-linking agents, and also the
10 above resins containing halogens and having a good flame-resistance.

(24) Cross-linkable acrylic resins deriving from substituted acrylates such as, for example, epoxy acrylates, urethane acrylates or polyester acryla-
15 tes.

(25) Alkyd resins, resins based on polyesters or acrylated resins cross-linked with melamine resins, urea resins, resins based on polyisocyanates or epoxy resins.

20 (26) Cross-linked epoxy resins deriving from polyepoxides such as, for example, bis-glycidyl ethers or cycloaliphatic diepoxides.

(27) Natural polymers such as, for example, cellulose, rubber, gelatine, and their derivatives chemically
25 modified to give homologous polymers such as, for

example, cellulose acetates, propionates and butyrates, or cellulose ethers such as, for example, methyl-cellulose; as well as hydrocarbon resins ("rosins") or their derivatives.

- 5 (28) Mixtures of the above polymers ("polyblends") such as, for example, PP/EPDM, polyamides/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTF/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastics PUR, PC/thermoplastics PUR, POM/acrylates, POM/MBS, PPO/HIPS, PPO/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPO.
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The compounds having formula (I) of the present invention are particularly useful in the stabilization of polycarbonates.

- 15 A further object of the present invention relates to polymeric compositions containing an organic polymer and an effective quantity of one or more compounds having general formula (I).

The compounds having general formula (I) of the present invention can be used as such or combined with other stabilizers, in the above polymeric compositions.

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In general, the above compounds having general formula (I) are used in a quantity ranging from about 0.1% to about 5% by weight of the weight of the polymeric compositions to be stabilized, although the

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quantity used varies according to the substrate to be stabilized and the final application. They are preferably added in a quantity ranging from about 0.5% to about 3% by weight of the weight of the polymeric
5 compositions to be stabilized.

The compounds having general formula (I), optionally in the presence of other additives, can be easily incorporated in the organic polymers to be stabilized using the conventional techniques. This incorporation
10 can take place before or during the formation of the end-product, for example, by mixing the compounds having general formula (I) in powder form with the polymer to be stabilized, or by adding these compounds to the polymer to be stabilized in the molten state or
15 in solution, or applying a solution or suspension of these compounds to the polymer to be stabilized, optionally evaporating the solvent used.

The elastomers can be stabilized as latexes. Another method for incorporating the compounds having
20 general formula (I) in the organic polymers comprises the addition of these before or during the polymerization of the corresponding monomers or before the cross-linking.

The compounds having general formula (I) or their
25 mixtures, can be added to the polymer to be stabilized

also in masterbatch form which comprises these compounds in a concentration ranging, for example, from 2.5% to 25% by weight.

The compounds having general formula (I) can be conveniently incorporated in the organic polymers to be stabilized by means of the following methods:

- in the form of emulsion or suspension (for example, in the case of latexes or polymers in emulsion);
- 10 - as a mixture of powders in the case of the conventional addition of additional compounds or mixtures of organic polymers;
- directly adding to the apparatus used for processing the organic polymers (for example, extruders, internal mixers, etc.);
- 15 - in the form of solution or molten product.

The polymeric compositions stabilized as said above can be converted to end-products such as, for example, fibers, films, tapes, sheets, multi-layer sheets, containers, tubes and other forms, by means of methods known in the art such as, for example, casting, hot moulding, spinning, extrusion or injection moulding.

The present invention therefore also relates to the use of the above polymeric compositions for the production of end-products.

The use of multi-layer systems is also of interest, in which one of the above compositions having a relatively high content of a compound having general formula (I), for example, between 5% and 15% by weight, is applied in the form of a thin film (10-100 μm in thickness) to a shaped article consisting of a polymer not containing or containing a small quantity of a compound having general formula (I). This application can be carried out during the formation of said article, for example, by means of a co-extrusion. The application can also be effected however on the end shaped-article, for example, by lamination with a film or by coating with a solution. The surface layer or layers of the end-article act as a UV filter which protects the inside of the articles from the deteriorating action of UV light. The upper layer preferably contains from 5% to 15% by weight, more preferably from 5% to 10% by weight, of at least one compound having general formula (I).

The use of the above compositions for the production of multi-layer systems, in which the upper layer having a thickness of 10-100 μm comprises these compositions, whereas the internal layer does not contain or contains a small quantity of a compound having general formula (I) consequently represents a further object of

the present invention.

The polymers stabilized as described above have a high resistance to degradation caused by atmospheric agents, in particular a high resistance to UV light.
5 They are therefore capable of maintaining their colour and brightness for a long period even when exposed to external agents.

The compositions described above can also be used as compositions for coating or painting ("coating
10 compositions") such as, for example, paints, lacquers, plastic-based compositions.

For the purposes of the present invention, coating or painting compositions are preferred in which the organic polymer is selected from:

- 15 (a) a thermoplastic polymer selected from thermoplastic polymers containing heteroatoms, in particular nitrogen, sulfur and/or oxygen, in the main chain, styrene copolymers, grafted styrene polymers and polymethyl methacrylates (PMMA); or
- 20 (b) a paint ligand.

Specific examples of thermoplastic polymers (a) containing heteroatoms, in particular nitrogen, sulfur and/or oxygen, in the main chain, are listed above under points 13 to 20. Among these, polycarbonates,
25 polyesters, polyamides, polyacetals, polyphenylene

oxides and polyphenylene sulfides are preferred; particularly preferred are polycarbonates, polyesters such as, for example, polyethylene terephthalate (PET), and polyamides (PA) such as, for example, PA 6 and PA 6/6; even more preferred are polycarbonates.

Specific examples of styrene copolymers and grafted styrene polymers (a) are listed above under points 6 and 7.

Paint ligands (b) can comprise at least one of the organic polymers specified herebelow. Specific examples of paints containing specific ligands are:

1. paints based on alkyd resins, acrylic resins, polyester resins, epoxy resins or melamine resins, which can be cross-linked at a low or high temperature, or mixtures of these resins, to which a cross-linking agent is optionally added;
2. polyurethane paints with two components based on acrylic resins containing hydroxyl groups, polyester resins or polyether resins and aliphatic or aromatic isocyanates, isocyanurates or polyisocyanates;
3. polyurethane paints with one component based on block isocyanates, isocyanurates or polyisocyanates which are unblocked during oven treatment;
4. paints with two components based on (poly)ketoi-

- mines and aliphatic or aromatic isocyanates, isocyanurates or polyisocyanates;
5. paints with two components based on (poly)keto-
imines and an unsaturated acrylic resin or a
5 polyacetoacetate resin or a methyl methacrylamidoglycolate;
6. paints with two components based on polyacrylates containing a carboxylic group or an amine group and polyepoxides;
- 10 7. paints with two components based on acrylic resins containing an anhydride group and a polyhydroxyl or polyamine compound;
8. paints with two components based on (poly)-oxazoline and acrylic resins containing an anhydride
15 group or unsaturated acrylic resins or aliphatic or aromatic isocyanates, or isocyanurates or polyisocyanates;
9. paints with two components based on unsaturated polyacrylates and polymalonates;
- 20 10. thermoplastic polyacrylic paints based on thermoplastic acrylic resins or non-self-crosslinking acrylic resins combined with etherified melamine resins;
11. systems for paints based on siloxane-modified
25 acrylic resins;

12. systems for paints based on fluoro-modified acrylic resins; and

13. systems for paints based on allyl glycidyl ethers.

The paints can be applied as one or two layers of
5 coating ("one- or two-coat") and the stabilizing compounds having formula (I) are preferably added to the upper colourless coating.

The paints can be applied to the substrate (metal, plastic, wood, etc.) using the conventional methods
10 such as, for example, brushing, spraying, pouring, dipping or electrophoresis.

A preferred embodiment of the present invention consists in paints or coatings (for example car coatings) comprising at least one compound having general
15 formula (I).

Ligands which can be used for the purpose are, for example, those listed above.

The compounds having general formula (I) of the present invention can be combined, as already mentioned
20 above, with other conventional additives or their mixtures. These additives are added in a quantity ranging from about 0.1% to about 5% by weight of the weight of the polymeric compositions to be stabilized, preferably from about 0.5% to about 3% by weight. Some
25 of the additives used are listed below as an example.

1. Antioxidants

1.1 Alkylated monophenols such as, for example:

- 2,6-di-t-butyl-4-methylphenol;
2-t-butyl-4,6-dimethylphenol;
5 2,6-di-t-butyl-4-ethylphenol;
2,6-di-t-butyl-4-n-butylphenol;
2,6-di-t-butyl-4-isobutylphenol;
2,6-di-cyclopentyl-4-methylphenol;
2-(α -methylcyclohexyl)-4,6-dimethylphenol;
10 2,6-dioctadecyl-4-methylphenol;
2,4,6-tricyclohexylphenol;
2,6-di-t-butyl-4-methoxymethylphenol;
2,6-di-nonyl-4-methylphenol;
2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol;
15 2,4-dimethyl-6-(1'-methylhectadec-1'-yl)phenol;
2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol; and
their mixtures.

1.2 Alkylthiomethylphenols such as, for example:

- 2,4-dioctylthiomethyl-6-t-butylphenol;
20 2,4-dioctylthiomethyl-6-methylphenol;
2,4-dioctylthiomethyl-6-ethylphenol;
2,6-didodecylthiomethyl-4-nonylphenol.

1.3 Hydroquinones and alkylated hydroquinones such as,
for example:

- 25 2,6-di-t-butyl-4-methoxyphenol;

- 2,5-di-t-butylhydroquinone;
2,5-di-t-amylhydroquinone;
2,6-diphenyl-4-octadecyloxyphenol;
2,6-di-t-butylhydroquinone;
5 2,5-di-t-butyl-4-hydroxyanisol;
3,5-di-t-butyl-4-hydroxyanisol;
3,5-di-t-butyl-4-hydroxyphenyl stearate;
bis(3,5-di-t-butyl-4-hydroxyphenyl)adipate.
- 1.4 Tocopherols such as, for example:
10 α -tocopherol, β -tocopherol, γ -tocopherol,
 δ -tocopherol and their mixtures (Vitamin E).
- 1.5 Hydroxylated thiophenyl ethers such as, for
example:
15 2,2'-thiobis-(6-t-butyl-4-methylphenol);
2,2'-thiobis-(4-octylphenol);
4,4'-thiobis-(6-t-butyl-3-methylphenol);
4,4'-thiobis-(6-t-butyl-2-methylphenol);
4,4'-thiobis-(3,6-di-sec-amylphenol);
4,4'-bis-(2,6-dimethyl-4-hydroxyphenyl)disulfide.
- 20 1.6 Alkylidene-bisphenols such as, for example:
2,2'-methylenebis-(6-t-butyl-4-methylphenol);
2,2'-methylenebis-(6-t-butyl-4-ethylphenol);
2,2'-methylenebis[4-methyl-6-(α -methylcyclohexyl)-
phenol];
25 2,2'-methylenebis(4-methyl-6-cyclohexylphenol);

- 2,2'-methylenebis(6-nonyl-4-methylphenol);
2,2'-methylenebis(4,6-di-t-butylphenol);
2,2'-ethylidenebis(4,6-di-t-butylphenol);
2,2'-ethylidenebis(6-t-butyl-4-isobutylphenol);
5 2,2'-methylenebis[6-(α -methylbenzyl)-4-nonylphenol];
2,2'-methylenebis[6-(α,α -dimethylbenzyl)-4-nonylphenol];
4,4'-methylenebis(2,6-di-t-butylphenol);
10 4,4'-methylenebis(6-t-butyl-2-methylphenol);
1,1-bis-(5-t-butyl-4-hydroxy-2-methylphenyl)-butane;
2,6-bis-(3-t-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol;
15 1,1,3-tris-(5-t-butyl-4-hydroxy-2-methylphenyl)-butane;
1,1-bis-(5-t-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane;
ethyleneglycol bis[3,3-bis(3'-t-butyl-4'-hydroxyphenyl)butyrate];
20 bis(3-t-butyl-4-hydroxy-5-methylphenyl)dicyclopentadiene;
bis[2-(3'-t-butyl-2'-hydroxy-5'-methylbenzyl)-6-t-butyl-4-methylphenyl]terephthalate;
25 1,1-bis(3,5-dimethyl-2-hydroxyphenyl)butane;

2,2-bis(3,5-di-t-butyl-4-hydroxyphenyl)propane;
2,2-bis(5-t-butyl-4-hydroxy-2-methylphenyl)-4-n-
dodecylmercaptobutane;
1,1,5,5-tetra(5-t-butyl-4-hydroxy-2-methylphenyl)-
5 pentane.

1.7 Benzyl compounds containing O, N or S such as, for
example:

3,5,3',5'-tetra-t-butyl-4,4'-dihydroxydibenzyl
ether;
10 octadecyl-4-hydroxy-3,5-dimethylbenzylmercapto-
acetate;
tris(3,5-di-t-butyl-4-hydroxybenzyl)amine;
bis(4-t-butyl-3-hydroxy-2,6-dimethylbenzyl)dithio-
terephthalate;
15 bis(3,5-di-t-butyl-4-hydroxybenzyl)sulfide;
iso-octyl-3,5-di-t-butyl-4-hydroxybenzylmercapto-
acetate;

1.8 Hydroxybenzylated malonates such as, for example:
dioctadecyl-2,2-bis(3,5-di-t-butyl-2-hydroxyben-
20 zyl)malonate;
dioctadecyl-2-(3-t-butyl-4-hydroxy-5-methylben-
zyl)malonate;
didodecylmercaptoethyl-2,2-bis(3,5-di-t-butyl-4-
hydroxybenzyl)malonate;
25 bis[4-(1,1,3,3-tetramethylbutyl)phenyl]-2,2-

bis(3,5-di-t-butyl-4-hydroxybenzyl)malonate.

1.9 Aromatic hydroxybenzyl compounds such as, for example:

1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene;

1,4-bis-(3,5-di-t-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene;

2,4,6-tris(3,5-di-t-butyl-4-hydroxybenzyl)phenol.

1.10 Triazine compounds such as, for example:

2,4-bis(octylmercapto)-6-(3,5-di-t-butyl-4-hydroxyaniline)-1,3,5-triazine;

2-octylmercapto-4,6-bis(3,5-di-t-butyl-4-hydroxyaniline)-1,3,5-triazine;

2-octylmercapto-4,6-bis(3,5-di-t-butyl-4-hydroxyphenoxy)-1,3,5-triazine;

2,4,6-tris-(3,5-di-t-butyl-4-hydroxyphenoxy)-1,2,3-triazine;

1,3,5-tris(3,5-di-t-butyl-4-hydroxybenzyl)isocyanurate;

1,3,5-tris(4-t-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate;

2,4,6-tris-(3,5-di-t-butyl-4-hydroxyphenylethyl)-1,3,5-triazine;

1,3,5-tris(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hexahydro-1,3,5-triazine;

1,3,5-tris-(3,5-dicyclohexyl-4-hydroxybenzyl)
isocyanurate.

1.11 Benzylphosphonates such as, for example:

dimethyl-2,5-di-t-butyl-4-hydroxybenzylphosphona-
5 te;

diethyl-3,5-di-t-butyl-4-hydroxybenzylphosphonate;

dioctadecyl-3,5-di-t-butyl-4-hydroxybenzylphospho-
nate;

dioctadecyl-5-t-butyl-4-hydroxy-3-methylbenzylpho-
10 sphonate;

calcium salts of monoethyl ester of 3,5-di-t-
butyl-4-hydroxybenzylphosphonic acid.

1.12 Acylaminophenols such as, for example:

4-hydroxylauranilide;

15 4-hydroxystearanilide;

octyl-N-(3,5-di-t-butyl-4-hydroxyphenyl)carbamate.

1.13 Esters of β -(3,5-di-t-butyl-4-hydroxyphenyl)pro-
pionic acid with monohydric or polyhydric alcohols
such as, for example:

20 methanol, ethanol, octanol, octadecanol, 1,6-he-
xandiol, 1,9-nonandiol, ethylene glycol, 1,2-pro-
panediol, neopentyl glycol, thiodiethylene glycol,
diethylene glycol, triethylene glycol, pentaeryth-
ritol, tris(hydroxyethyl) isocyanurate, N,N'-
25 bis(hydroxyethyl)oxamide, 3-thioundecanol, 3-

thiopentadecanol, trimethylhexandiol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

1.14 Esters of β -(5-t-butyl-4-hydroxy-3-methylphenyl)propionic acid with monohydric or polyhydric alcohols such as, for example:

methanol, ethanol, octanol, octadecanol, 1,6-hexandiol, 1,9-nonandiol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexandiol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

1.15 Esters of β -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with monohydric or polyhydric alcohols such as, for example:

methanol, ethanol, octanol, octadecanol, 1,6-hexandiol, 1,9-nonandiol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexandiol, trimethylol-

propane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

1.16 Esters of (3,5-di-*t*-butyl-4-hydroxyphenyl)acetic acid with monohydric or polyhydric alcohols such as, for example:

methanol, ethanol, octanol, octadecanol, 1,6-hexandiol, 1,9-nonandiol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, *N,N'*-bis(hydroxyethyl)oxamide, 3-thioundecanol, 3-thiopentadecanol, trimethylhexandiol, trimethylolpropane, 4-hydroxymethyl-1-phospho-2,6,7-trioxabicyclo[2.2.2]octane.

1.17 Amides of β -(3,5-di-*t*-butyl-4-hydroxyphenyl)propionic acid such as, for example:

N,N'-bis(3,5-di-*t*-butyl-4-hydroxyphenylpropionyl)hexamethylenediamine;

N,N'-bis(3,5-di-*t*-butyl-4-hydroxyphenylpropionyl)trimethylenediamine;

N,N'-bis(3,5-di-*t*-butyl-4-hydroxyphenylpropionyl)hydrazine.

2. Ultra-violet ray and light stabilizers.

2.1 Derivatives of 2-(2'-hydroxyphenyl)benzotriazoles

such as, for example:

- 2-(2'-hydroxy-5'-methylphenyl)benzotriazole;
2-(3',5'-di-t-butyl-2'-hydroxyphenyl)benzotriazole;
2-(5'-t-butyl-2'-hydroxyphenyl)benzotriazole;
5 2-[2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl]benzotriazole;
2-(3',5'-di-t-butyl-2'-hydroxyphenyl)-5-chlorobenzotriazole;
2-(3'-t-butyl-2'-hydroxy-5'-methylphenyl)-5-chlorobenzotriazole;
10 2-(3'-sec-butyl-5'-t-butyl-2'-hydroxyphenyl)benzotriazole;
2-(2'-hydroxy-4'-octyloxyphenyl)-benzotriazole;
2-(3',5'-di-t-amyl-2'-hydroxyphenyl)-benzotriazole;
15 2-[3',5'-bis(α,α -dimethylbenzyl)-2'-hydroxyphenyl]benzotriazole;
mixtures of 2-[3'-t-butyl-2'-hydroxy-5'-(2-octyloxy-carbonyl-ethyl)phenyl]-5-chlorobenzotriazole, 2-[3'-t-butyl-5'-(2-(2-ethylhexyloxy)carbonyl-ethyl)-2'-hydroxyphenyl]-5-chlorobenzotriazole, 2-[3'-t-butyl-2'-hydroxy-5'-(2-methoxycarbonyl-ethyl)phenyl]-5-chlorobenzotriazole, 2-[3'-t-butyl-2'-hydroxy-5'-(2-methoxycarbonyl-ethyl)phenyl]benzotriazole, 2-[3'-t-butyl-2'-hydroxy-5'-(2-octylo-
- 20
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5 xycarbonylethyl)phenyl]benzotriazole, 2-[3'-t-butyl-5'-(2-(2-ethylhexyloxy)carbonylethyl)-2'-hydroxyphenyl]benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole and 2-[3'-t-butyl-2'-hydroxy-5'-(2-iso-octyloxycarbonyl-ethyl)phenyl]benzotriazole, 2,2'-methylene-bis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazol-2-yl-phenol]; esterification product of 2-[3'-t-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-10 benzotriazole with polyethylene glycol 300;
 $[R-CH_2CH_2-COO(CH_2)_3]_2$ wherein $R = 3'-t\text{-butyl-4-hydroxy-5'-2H-benzotriazol-2-yl-phenyl}$.

2.2 Derivatives of 2-hydroxybenzophenones such as, for example: 4-hydroxy-; 4-methoxy-; 4-octyloxy-; 4-decyloxy-; 4-dodecyloxy-; 4-benzyloxy-; 4,2',4'-15 trihydroxy-; 2'-hydroxy-4,4'-dimethoxy.

2.3 Esters of benzoic acids, optionally substituted, such as, for example: phenyl salicylate, 4-t-butylphenyl salicylate, octylphenyl salicylate, 20 benzoyl-resorcinol, bis(4-t-butylbenzoyl)-resorcinol, dibenzoyl-resorcinol, 2,4-di-t-butylphenyl-3,5-di-t-butyl-4-hydroxybenzoate, hexadecyl-3,5-di-t-butyl-4-hydroxybenzoate, octadecyl-3,5-di-t-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-t-butyl-phenyl-3,5-di-t-butyl-4-hydroxybenzoate. 25

- 2.4 Acrylates such as, for example, ethyl or isooctyl α -cyano- β , β -diphenylacrylate; methyl α -carbomethoxycinnamate, methyl or butyl α -cyano- β -methyl-p-methoxycinnamate, methyl α -carbomethoxy-p-methoxycinnamate, N-(β -carbomethoxy- β -cyanovinyl)-2-methylindoline.
- 2.5 Nickel compounds such as, for example, complexes of 2,2'-thio-bis-[4-(1,1,3,3-tetramethylbutyl)phenol], for example 1:1 or 1:2 complexes, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of monoalkyl esters of 4-hydroxy-3,5-di-t-butylbenzyl-phosphonic acid, such as methyl or ethyl esters, nickel complexes with ketoximes such as 2-hydroxy-4-methylphenyl undecyl ketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazol with or without additional ligands.
- 2.6 Sterically hindered amines such as, for example: bis(2,2,6,6-tetramethyl-piperidyl)sebacate; bis(2,2,6,6-tetramethyl-piperidyl)succinate; poly-methylpropyl-3-oxy[4-(2,2,6,6-tetramethyl)piperidinyl]siloxane; bis(1,2,2,6,6-pentamethyl-piperidyl)sebacate; bis(1,2,2,6,6-pentamethyl-piperidyl)n-butyl-3,5-

- di-t-butyl-4-hydroxybenzylmalonate;
condensation product between 1-(2-hydroxyethyl)-
2,2,6,6-tetra-4-hydroxypiperidine and succinic
acid;
- 5 condensation product between N,N'-bis(2,2,6,6-
tetramethyl-4-piperidyl)hexamethylenediamine and
4-t-octylamino-2,6-dichloro-1,3,5-triazine;
tris(2,2,6,6-tetramethyl-4-piperidyl)nitrilotri-
acetate;
- 10 tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-
butane-tetracarboxylate;
1,1'-(1,2-ethanodiyl)bis(3,3,5,5-tetramethyl-
piperazinone;
4-benzoyl-2,2,6,6-tetramethylpiperidine;
- 15 4-stearyloxy-2,2,6,6-tetramethylpiperidine;
bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-
(2-hydroxy-3,5-di-t-butylbenzyl)malonate;
3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspi-
ro[4.5]decan-2,4-dione;
- 20 bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)se-
bacate;
bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)suc-
cinate;
- 25 condensation product between N,N'-bis(2,2,6,6-
tetramethyl-4-piperidyl)hexamethylenediamine and

- 4-morpholine-2,6-dichloro-1,3,5-triazine;
condensation product between 2-chloro-4,6-di-(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane;
- 5 condensation product between 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane;
- 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decano-2,4-dione;
- 10 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyrrolidin-2,5-dione;
- 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidin-2,5-dione.
- 2.7 Oxamides such as, for example:
- 15 4,4'-dioctyloxyoxanilide;
- 2,2'-diethoxyoxanilide;
- 2,2'-dioctyloxy-5,5'-di-t-butoxanilide;
- 2,2'-didodecyloxy-5,5'-di-t-butoxanilide;
- 2-ethoxy-2'-ethyloxanilide;
- 20 N,N'-bis(3-dimethylaminopropyl)oxamide;
- 2-ethoxy-5-t-butyl-2'-ethoxanilide and its mixtures with 2-ethoxy-2'-ethyl-5,4'-di-t-butoxanilide; and mixtures of disubstituted ortho- and paramethoxy anilides and mixtures of disubstituted
- 25 ortho and para-ethoxy anilides.

2.8 2-(2-hydroxyphenyl)-1,3,5-triazines such as, for example:

2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine;

5 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine;

2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine;

10 2,4-bis-(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine;

2-(2-hydroxy)-4,6-bis(4-methylphenyl)-1,3,5-triazine;

2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine;

15 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine;

2-[2-hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine.

3. "Metal-deactivators" such as, for example: N,N-

20 diphenyloxamide, N-salicylal-N'-salicyloyl-hydrazine, N,N'-bis(salicyloyl)hydrazine; N,N'-bis(3,5-di-t-butyl-4-hydroxyphenylpropionyl)hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diace-

25

tyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.

4. Phosphites and phosphonites such as, for example:
- 5 triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl)phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-t-butylphenyl)phosphite, diisodecyl
- 10 pentaerythritol diphosphite, bis(2,4-di-t-butylphenyl)pentaerythritol diphosphite, bis(2,5-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-t-butyl-6-methylphenyl)pentaerythritol diphosphite, bis[2,4,5-tris(t-butylphenyl)]pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis-(2,4-di-t-butyl-phenyl)-4,4'-diphenylilenediphosphonite, 5-iso-octyloxy-2,4,8,10-tetra-t-butyl-12H-di-benzo[d,g]-1,3,2-dioxaphosphocine, 6-
- 20 fluoro-2,4,8,10-tetra-t-butyl-12-methyl-dibenzo[d,g]-1,3,2-dioxaphosphocine, bis(2,4-di-t-butyl-6-methylphenyl)methylphosphite, bis(2,4-di-t-butyl-6-methylphenyl)ethylphosphite.
5. Agents which are capable of destroying peroxides
- 25 such as, for example, esters of β -thiodipropionic

- acid such as lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyldisulfide pentaerythritol tetrakis
- 5 (β -dodecylmercapto)propionate.
6. Stabilizers of polyamides such as, for example, copper salts combined with compounds of iodine and/or phosphorous, divalent manganese salts.
7. Basic co-stabilizers such as, for example: melamine,
- 10 polyvinylpyrrolidone, dicyanodiamide, triallyl cyanurate, derivatives of urea, derivatives of hydrazine, amines, polyamides, polyurethanes, salts of alkaline metals and salts of earth-alkaline metals of fatty acids such as, for
- 15 example, Ca-stearate, Zn-stearate, Mg-stearate, Mg-behenate, Na-ricinoleate, K-palmitate, antimony-pyrocatecholate, tin-pyrocatecholate.
8. Nucleating agents such as, for example: 4-t-butylbenzoic acid, adipic acid, diphenylacetic acid.
- 20 9. Fillers and reinforcing agents such as, for example: calcium carbonate, silicates, glass fibres, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite.
- 25 10. Other additives such as, for example: plastici-

- zers, lubricants, emulsifying agents, pigments, optical brighteners, flame-retardants (for example, bromurates, chlorurates, phosphorates and phosphorous/halogen mixtures), antistatic agents, blowing agents, thiosynergizing agents such as, for example, dilauryl thiodipropionate or distearyl thiodipropionate.
- 5
11. Benzofuranones and indolinones such as, for ex.:
3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-t-butylbenzofuran-2-one;
10 5,7-di-t-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one;
3,3'-bis[5,7-di-t-butyl-3-[4-(2-hydroxyethoxy)phenyl]benzofuran-2-one];
15 5,7-di-t-butyl-3-(4-ethoxyphenyl)benzofuran-2-one;
3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-t-butylbenzofuran-2-one;
3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-t-butylbenzofuran-2-one;
20 or those described in U.S. patents 4.325.863, 4.338.244, 5.175.312, 5.216.052, 5.252.643, 4.316.611, 4.316.622, 4.316.876 or in European patent applications 589.839 and 591.102.

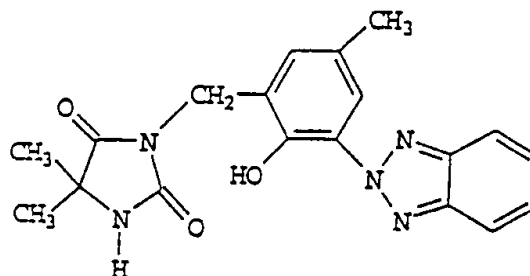
Some illustrative but non-limiting examples are
25 provided hereunder for a better understanding of the

present invention and for its embodiment.

EXAMPLE 1

Preparation of Compound Nr. 1 having the formula:

5



15 g (0.067 moles) of 2-(2'-hydroxy-5-methyl-
10 phenyl)-2H-benzotriazole, 7.99 g (0.079 moles) of di-n-propylamine, 3.36 g (0.112 moles) of para-formaldehyde and 30 ml of n-butanol are charged into a 3-necked 100 ml flask, equipped with a drip funnel, thermometer and condenser. The mixture is heated to reflux temperature
15 for 44 hours and the reaction is controlled by thin layer chromatography (T.L.C.) using hexane:ethyl acetate in a ratio 1:1 as mobile phase.

At the end of the reaction, the solvent is eliminated by evaporation obtaining the Mannich base corresponding to 2-(2'-hydroxy-3-dipropylaminomethyl-5-methylphenyl)-2H-benzotriazole.
20

20 g (0.059 moles) of 2-(2'-hydroxy-3-dipropylaminomethyl-5-methylphenyl)-2H-benzotriazole, 7.6 g (0.059 moles) of 5,5-dimethyl-2,4-imidazolidinedione
25 and 75 ml of 1,2,4-trimethylbenzene are subsequently

charged into a 3-necked 250 ml flask, equipped with a thermometer and condenser. After heating the mixture until the complete dissolution of the reagents, 0.16 g of sodium methyllate are added as catalyst, the temperature is brought to 168°C in a stream of nitrogen and the mixture is maintained at this temperature for 30 hours.

At the end of the reaction, which is controlled by thin layer chromatography (T.L.C.) as described above, the mixture is cooled to room temperature, the catalyst is eliminated by filtration and, after evaporation of the solvent, the desired compound is crystallized in the presence of 2-methoxyethanol obtaining 18 g of a white powder corresponding to Compound Nr. 1 having the following characteristics:

- melting point (DSC): 216°C.
- I.R. in nujols (cm⁻¹): 3100, 1770, 1714
- ¹H-NMR (200 MHz, CDCl₃) δ (ppm): 1.47 (s, 6H); 2.36 (s, 3H); 4.84 (s, 2H); 5.88 (brs, 1H); 7.06 (d, J = 1.6 Hz, 1H); 7.45 (dd, 2H); 7.89 (dd, 2H); 8.13 (d, J = 1.6 Hz, 1H); 11.5 (s, 1H).
- ¹³C-NMR (50 MHz, CDCl₃) δ (ppm): 20.6; 25.19; 37.58; 58.84; 117.63; 120.83; 124.97; 127.70; 129.00; 130.45; 142.80; 145.60; 156.20; 176.90.

Elemental analysis:

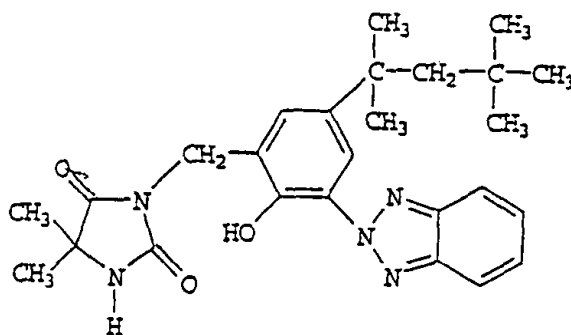
=====			
ELEMENTAL		C ₁₉ H ₁₉ N ₅ O ₃	
ANALYSIS	C	H	N

5	CALCULATED	62.47%	5.21% 19.17%
	FOUND	62.40%	5.20% 19.14%
=====			

EXAMPLE 2

Preparation of Compound Nr. 2 having the formula:

10



15

15 g (0.046 moles) of 2-(2'-hydroxy-5-t-octyl-phenyl)-2H-benzotriazole, 5.45 g (0.054 moles) of di-n-propylamine, 23 g (0.077 moles) of para-formaldehyde and 30 ml of n-butanol are charged into a 3-necked 100 ml flask, equipped with a drip funnel, thermometer and condenser. The mixture is heated to reflux temperature for 44 hours and the reaction is controlled by thin layer chromatography (T.L.C.) using hexane:ethyl acetate in a ratio 1:1 as mobile phase.

25 At the end of the reaction, the solvent is elimi-

nated by evaporation obtaining the Mannich base corresponding to 2-(2'-hydroxy-3-dipropylaminomethyl-5-t-octylphenyl)-2H-benzotriazole.

20 g (0.046 moles) of 2-(2'-hydroxy-3-dipropyl-
5 aminomethyl-5-t-octylphenyl)-2H-benzotriazole, 5.9 g
(0.046 moles) of 5,5-dimethyl-2,4-imidazolidinedione
and 75 ml of 1,2,4-trimethylbenzene are subsequently
charged into a 3-necked 250 ml flask, equipped with a
thermometer and condenser. After heating the mixture
10 until the complete dissolution of the reagents, 0.15 g
of sodium methylate are added as catalyst, the tempera-
ture is brought to 168°C in a stream of nitrogen and
the mixture is maintained at this temperature for 25
hours.

15 At the end of the reaction, which is controlled by
thin layer chromatography (T.L.C.) as described above,
the mixture is cooled to room temperature, the catalyst
is eliminated by filtration and, after evaporation of
the solvent, the desired compound is crystallized in
20 the presence of methanol obtaining 19.5 g of a white
powder corresponding to Compound Nr. 2 having the
following characteristics:

- melting point (DSC): 130°C.
- ¹H-NMR (200 MHz, CDCl₃) δ (ppm): 0.8 (s, 9H); 1.37
25 (s, 6H); 1.48 (s, 6H); 1.70 (s, 2H); 4.85 (s, 2H);

5.91 (brs, 1H); 7.05 (d, J = 1.6 Hz, 1H); 7.45 (dd, 2H); 7.9 (dd, 2H); 8.3 (d, J = 1.6 Hz, 1H); 11.6 (s, 1H).

- ¹³C-NMR (50 MHz, CDCl₃) δ (ppm): 25.0; 28.9; 31.0; 32.0; 37.8; 38.2; 56.6; 60.0; 117.6; 118.3; 124.0; 126.0; 127.0; 129.0; 142.2; 142.7; 144.8; 155.0; 177.0.

Elemental analysis:

10	ELEMENTAL	C ₂₆ H ₃₃ N ₅ O ₃		
	ANALYSIS	C	H	N

	CALCULATED	67.39%	7.13%	15.00%
	FOUND	67.26%	7.07%	14.90%

15 =====

EXAMPLE 3

Ultraviolet ray absorption data

10⁻² M solutions in chloroform of compounds Nr. 1 and 2 obtained as described above and Tinuvin 900 produced and sold by Ciba Geigy, are prepared. These solutions are analyzed using a diode spectrophotometer of the type "hp 8452A DIODE ARRAY spectrophotometer" and the ε molar extinction coefficients at λ = 400 nm are indicated in Table 1.

TABLE 1

=====		
	COMPOUND	ϵ (l.cm ⁻¹ .mol ⁻¹)
		(400 nm)
5	Nr. 1	40
	Nr. 2	46
	TINUVIN 900	180

=====

EXAMPLE 4

10 Thermogravimetric data

Using a standard instrument for thermogravimetric analysis (TA Instrument Model 3000 of Mettler) the following isothermal and gravimetric data are determined relating to Compounds Nr. 1 and Nr. 2 which are
15 compared with the data of Tinuvin 900 produced and sold by Ciba Geigy. The data are indicated in Table 2.

TABLE 2

=====				
COMPOUND	ISOTHERM AT 280°C;		SCANNING AT	
Nr.	18 Nl/h N ₂ ; TIME IN		10°C/min; 18 Nl/h	
5	MINUTES TO INDICATE		N ₂ ; TEMPERATURE IN	
	LOSS IN WEIGHT OF		°C TO INDICATE LOSS	
	THE STABILIZER		IN WT. OF STABILIZER	
	10%	50%	10%	50%
1	*	-	302	387
10 2	**	-	298	362
Tinuvin 900	***	20	281	319
=====				

*: after 20 minutes;

**: after 18 minutes;

15 ***: after 4.2 minutes.

EXAMPLE 5

Stabilization in polycarbonate

20 g of bisphenol A polycarbonate are dissolved in 100 ml of methylene chloride, at room temperature under stirring. After several hours, when the polycarbonate has completely dissolved, 0.1 g of Compound Nr. 1 or Compound Nr. 2 or Tinuvin 900 are added (addition equal to 0.5%). For comparative purposes a solution without light stabilizers is prepared.

25 Films with a thickness of 100 µm are prepared from

the above solutions by casting.

The films obtained are subjected to accelerated aging in an Atlas CI 65 Weatherometer under the following conditions:

- 5 - black panel temperature: 60°C;
- relative humidity: 50%.

Before beginning the aging and afterwards, at regular intervals, the yellow index (YI) of the above films is measured using the method ASTM E 313. The crushing time is also analyzed.

The results obtained are indicated in Table 3.

TABLE 3

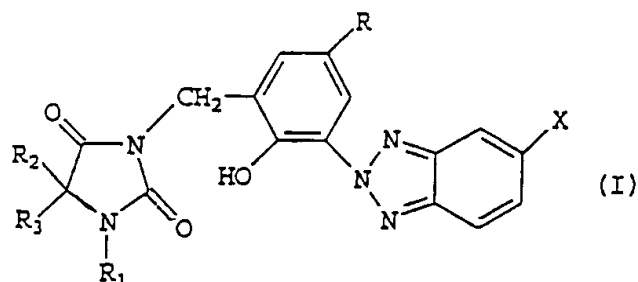
COMPOUND		EXPOSURE TIME						
15	Nr.	(h)						
		0	250	500	600	800	1000	1500
	-	0.4	4.0	12.4*	-	-	-	-
	1	0.3	3.0	5.0	8.0	9.5	10.6	12.6*
	2	0.5	3.6	5.8	8.7	9.3	10.9	12.7*
20	Tinuvin 900	0.4	3.2	6.0	9.5	10.8	12.0*	-

*: crushing of sample.

CLAIMS

1. 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I):

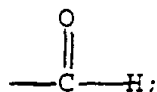
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wherein:

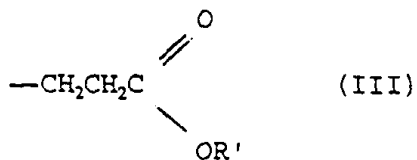
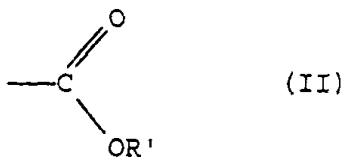
- X represents a hydrogen atom; a halogen atom selected from chlorine and bromine; a linear or branched C₁-C₁₈ alkyl group; a linear or branched C₁-C₁₈ alkoxy group; a cyano group;
- R represents a halogen atom selected from chlorine and bromine; a linear or branched C₁-C₁₈ alkyl group; a linear or branched C₂-C₁₈ alkenyl group; a linear or branched C₂-C₁₈ alkynyl group; a C₅-C₁₈ cycloalkyl group, said cycloalkyl group optionally substituted; a C₇-C₁₅ arylalkyl or alkylaryl group; a C₆-C₁₄ aryl group, said aryl group optionally substituted; a linear or branched C₁-C₁₈ alkoxy group; a heterocyclic group with 5 or 6 atoms containing at least one heteroatom selected from oxygen, nitrogen and sulfur, said heterocyclic group optionally substituted; a

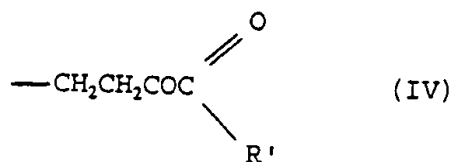
group having the formula:



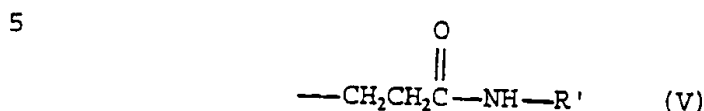
5 a -COR₄ group or a -NR₅R₆ group wherein R₄, R₅ and R₆, the same or different, represent a linear or branched C₁-C₁₈ alkyl group; a linear or branched C₂-C₁₈ alkenyl group; a linear or branched C₂-C₁₈ alkynyl group; a C₅-C₁₈ cycloalkyl group, said cycloalkyl group
10 optionally substituted; a C₇-C₁₅ arylalkyl or alkylaryl group; a C₆-C₁₄ aryl group, said aryl group optionally substituted; a heterocyclic group with 5 or 6 atoms containing at least one heteroatom selected from oxygen,
15 nitrogen and sulfur, said heterocyclic group optionally substituted;
or R represents an ester group having general formula (II), (III) or (IV):

20



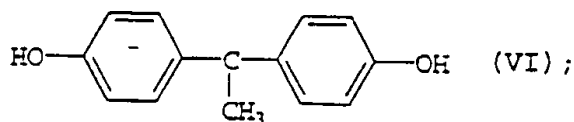


or an amide group having general formula (V):



wherein R' represents a linear or branched C₁-C₁₈ alkyl group; a linear or branched C₂-C₁₈ alkenyl group; a linear or branched C₂-C₁₈ alkynyl group; a C₃-C₁₈ cycloalkyl group, said cycloalkyl group optionally substituted; a C₇-C₁₅ arylalkyl or alkylaryl group; a C₆-C₁₄ aryl group, said aryl group optionally substituted; a linear or branched C₁-C₁₈ alkoxy group; a heterocyclic group with 5 or 6 atoms containing at least one heteroatom selected from oxygen, nitrogen and sulfur, said heterocyclic group optionally substituted;

or R represents a 4,4'-ethylidenebisphenol group having formula (VI):



R₁ represents a hydrogen atom; a linear or

branched C_1-C_{18} alkyl group; a linear or
 branched C_2-C_{18} alkenyl group; a linear or
 branched C_2-C_{18} alkynyl group; a C_5-C_{18} cyclo-
 alkyl group, said cycloalkyl group optionally
 5 substituted; a C_7-C_{15} arylalkyl or alkylaryl
 group; a C_6-C_{14} aryl group, said aryl group
 optionally substituted; a heterocyclic group
 with 5 or 6 atoms containing at least one
 heteroatom selected from oxygen, nitrogen and
 10 sulfur, said heterocyclic group optionally
 substituted; an acyl group having general
 formula (VII):



15 or an ester group having general formula
 (VIII):



20 wherein R' has the same meanings described
 above;

- R_2 and R_3 , the same or different, represent a
 hydrogen atom; a linear or branched C_1-C_{18}
 alkyl group; a phenyl group; a heterocyclic
 group with 5 or 6 atoms containing at least
 25 one heteroatom selected from oxygen, nitrogen

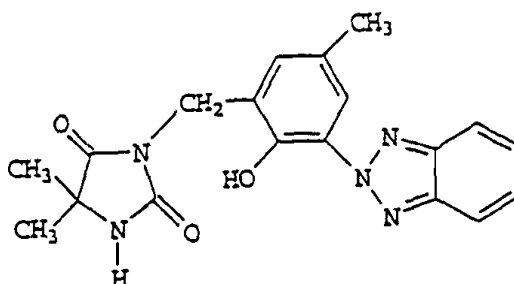
and sulfur, said heterocyclic group optionally substituted.

2. The 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to claim 1, wherein
5 the C₅-C₁₈ cycloalkyl groups, the C₆-C₁₄ aryl groups and the heterocyclic groups with 5 or 6 atoms are substituted with: halogen atoms selected from chlorine and bromine, linear or branched C₁-C₁₈ alkyl groups, linear or branched C₂-C₁₈ alkenyl groups;
10 groups; linear or branched C₂-C₁₈ alkynyl groups, OH groups, NH groups, SH groups.
3. The 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to claim 1, wherein the C₁-C₁₈ alkyl groups are: methyl, ethyl, propyl,
15 isopropyl, n-butyl, sec-butyl, t-butyl, t-amyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-dodecyl, 1,1,7,7-tetramethyloctyl, n-octadecyl.
4. The 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to claim 1, wherein
20 the C₂-C₁₈ alkenyl groups are: vinyl, propylene, butylene, pentylene, hexylene.
5. The 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to claim 1, wherein the C₂-C₁₈ alkynyl groups are: acetylene, propyne,
25 butyne, 2-butyne.

6. The 2-(2'-hydroxyphenyl)benzotriazoles having
general formula (I) according to claim 1, wherein
the C₇-C₁₈ cycloalkyl groups, optionally substituted,
are: cyclohexyl, cyclopentyl, methylcyclohe-
5 xyl.
7. The 2-(2'-hydroxyphenyl)benzotriazoles having
general formula (I) according to claim 1, wherein
the C₇-C₁₅ arylalkyl or alkylaryl groups are:
benzyl, 2-phenylethyl, 4-t-butylbenzyl.
- 10 8. The 2-(2'-hydroxyphenyl)benzotriazoles having
general formula (I) according to claim 1, wherein
the C₆-C₁₄ aryl groups, optionally substituted, are:
phenyl, naphthyl, anthracenyl, 2-hydroxyphenyl.
9. The 2-(2'-hydroxyphenyl)benzotriazoles having
15 general formula (I) according to claim 1, wherein
the C₁-C₁₈ alkoxyl groups are: methoxyl, ethoxyl,
propoxyl, n-butoxyl.
10. The 2-(2'-hydroxyphenyl)benzotriazoles having
general formula (I) according to claim 1, wherein
20 the heterocyclic groups with 5 or 6 atoms, option-
ally substituted, are: piperidine, morpholine,
piperazine, triazole, tetramethylpiperidine,
pentamethylpiperidine, tetramethylmorpholine,
pentamethylmorpholine, 4-hydroxy-tetramethylpi-
25 peridine.

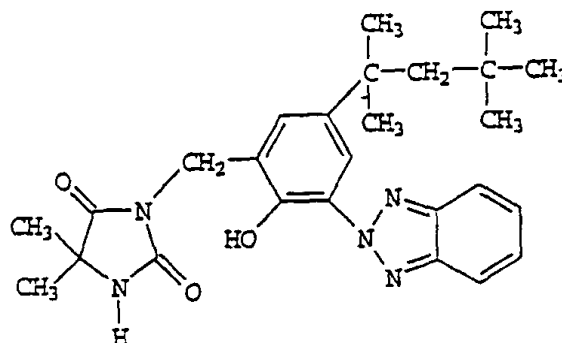
11. 2-(2'-hydroxyphenyl)benzotriazole according to claim 1, having the formula:

5



12. 2-(2'-hydroxyphenyl)benzotriazole according to claim 1, having the formula:

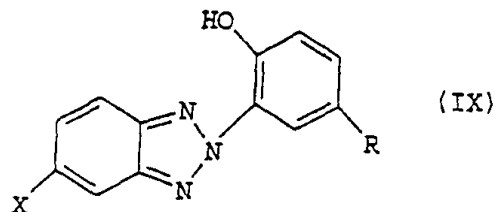
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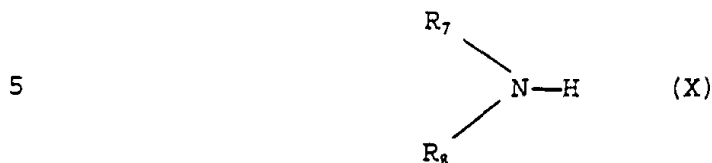
13. A process for the preparation of the 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to any of the previous claims, which comprises:

- 20 (a) reacting a 2-(2'-hydroxyphenyl)benzotriazole having general formula (IX):



25

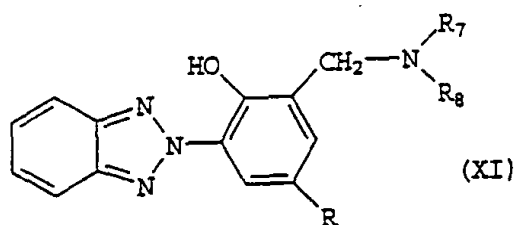
wherein X and R have the same meanings defined above, with a secondary amine having general formula (X):



wherein R_7 and R_8 , the same or different, preferably the same, represent a linear alkyl group having from 3 or more carbon atoms, in the presence of formaldehyde or para-formaldehyde and an inert organic solvent, at a temperature ranging from 95°C to 100°C, for a time ranging from 40 hours to 60 hours, obtaining the Mannich base having general formula (XI):

10

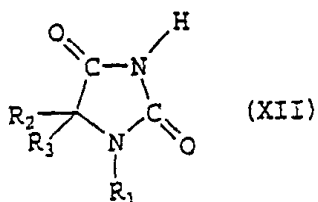
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20 wherein R, R_7 and R_8 have the same meanings defined above;

(b) reacting the Mannich base having general formula (XI) obtained in step (a) with a 2,4-imidazolidinedione having general formula (XII):

25



- 5 wherein R_1 , R_2 and R_3 have the same meanings described above, in the presence of an alkaline catalyst and an inert organic solvent having a boiling point at 160°C, at a temperature ranging from 20°C to 200°C, for a
- 10 time ranging from 20 hours to 30 hours, obtaining a solution from which, after cooling to room temperature and eliminating the catalyst by filtration and the solvent by evaporation, the desired compound is isolated
- 15 by crystallization in the presence of an inert organic solvent selected from alcohols; monoalkylethers; linear or cyclic aliphatic hydrocarbons; aromatic hydrocarbons; chlorinated aromatic solvents.
- 20 14. The process according to claim 13, wherein step (a) is carried out in the presence of para-formaldehyde and n-butanol.
15. The process according to claim 13, wherein the catalysts used in step (b) are: sodium methylate,
- 25 sodium hydroxide, potassium hydroxide, lithium

hydroxide.

16. The process according to claim 13, wherein the inert organic solvents having a boiling point at 160°C used in step (b) are: 1,2,4-trimethylbenzene, 4-isopropylbenzene.
17. The process according to claim 13, wherein step (b) is carried out at a temperature ranging from 40°C to 170°C.
18. The process according to claim 13, wherein the crystallization solvents used in step (b) are: methanol, 2-methoxyethanol, hexane, heptane, cyclohexane, methylcyclohexane, toluene, chlorobenzene.
19. Polymeric compositions containing an organic polymer and an effective quantity of one or more 2-(2'-hydroxyphenyl)benzotriazoles with general formula (I) according to any of the previous claims.
20. The polymeric compositions according to claim 19, wherein the 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) are combined with other stabilizers.
21. The polymeric compositions according to claim 19 or 20, wherein the organic polymer is selected from polycarbonates.
22. End-products obtained from the processing of the

polymeric compositions according to any of the claims from 19 to 21.

23. Use of the 2-(2'-hydroxyphenyl)benzotriazoles having general formula (I) according to any of the claims from 1 to 18, as light stabilizers for organic polymers.
24. Use of the polymeric compositions according to claim 19 or 20, for the production of multilayer systems, wherein the upper layer having a thickness of 10-100 μm comprises these compositions, whereas the internal layer does not contain or contains a small quantity of a compound having general formula (I) according to any of the claims from 1 to 18.
25. Use of the polymeric compositions according to claim 19 or 20, as coating or painting compositions.

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 98/06654

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C07D403/10 C08K5/3475

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C07D C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	EP 0 867 435 A (GREAT LAKES CHEMICAL ITALIA) 30 September 1998 see the whole document ---	1-25
Y	EP 0 057 160 A (CIBA GEIGY AG) 4 August 1982 see claim 1 ---	1-21
Y	US 4 077 971 A (FUJITA TAIRA ET AL) 7 March 1978 see claim 1 ---	1-21
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☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search

4 February 1999

Date of mailing of the international search report

12.02.99

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Authorized officer

Gettins, M

INTERNATIONAL SEARCH REPORT

Inter national Application No

PCT/EP 98/06654

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 4 044 019 A (SCHMIDT ANDREAS ET AL) 23 August 1977 see claim 1 ---	1-21
Y	US 3 629 192 A (HELLER HANSJORG ET AL) 21 December 1971 see claim 1 ---	1-21
Y	CH 476 745 A (J.R.GEIGY AG) 15 August 1969 see claim 1 ---	1-21
Y	FR 1 324 899 A (J.R.GEIGY AG) 22 July 1963 see claim 1 -----	1-21

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP 98/06654

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.: 22
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Claims Nos.: 22

Claim 22 reads "end-products obtained from the processing of polymeric compositions" wherein the polymer compositions are any organic polymers in combination with the compounds (I) of the current application. The scope of the claim is theoretically anything which can possibly be made from polymers. Apart from the fact that that this represents an unreasonable overgeneralisation of the specific uses (e.g. paints) given in the application it represents a functional definition of a physical entity and as such is not allowable. Furthermore since there is no indication as to how the said end-products can be made there is a lack of sufficient disclosure for claim 22.

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 98/06654

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